Experimental Evaluation of Mixed Fluid Reactions between Supercritical CO_2 and NaCl Brine: Relevance to Geologic Aquifer Carbon Sequestration

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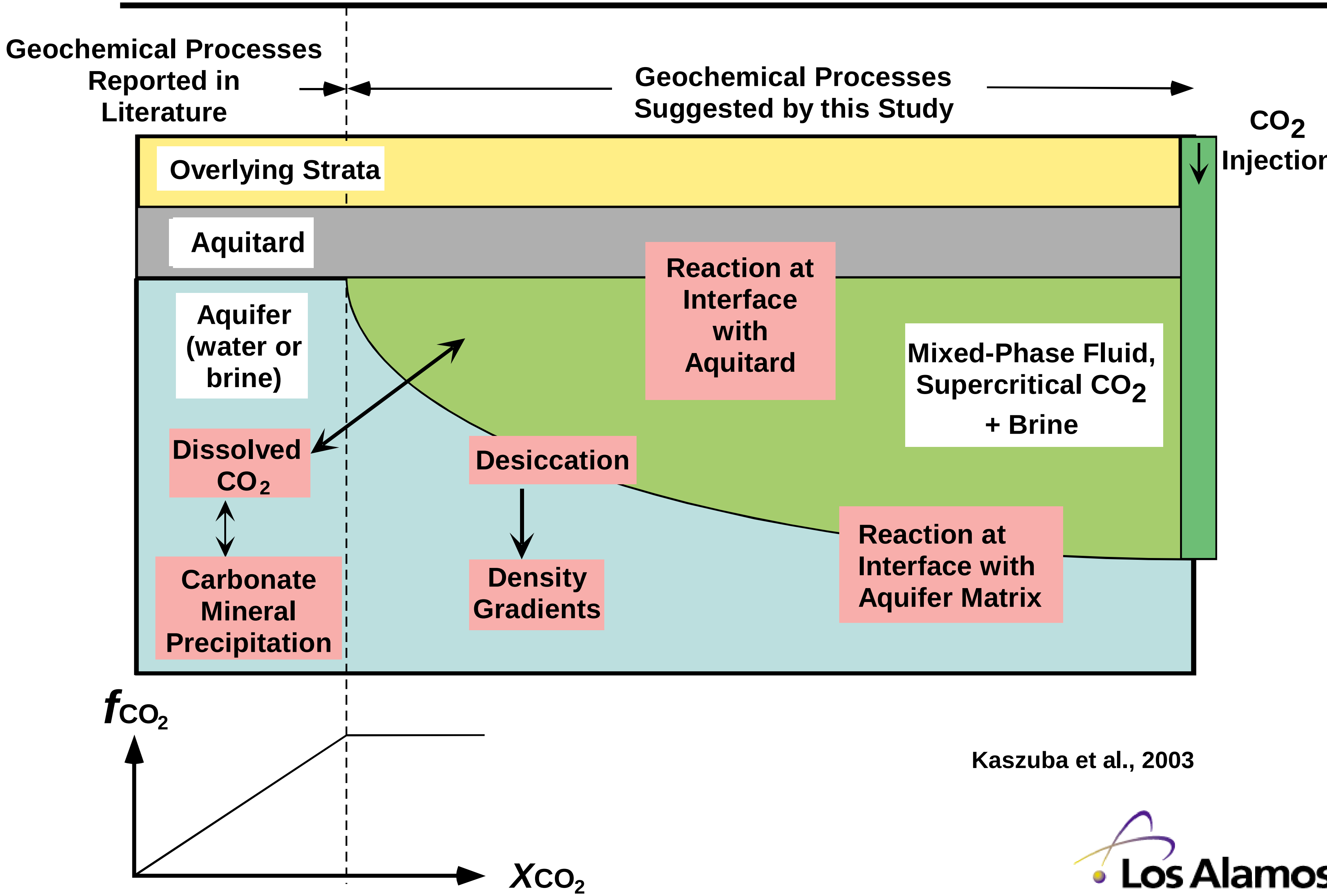
Background - Geologic Sequestration of CO_2



Kaszuba et al., 2003

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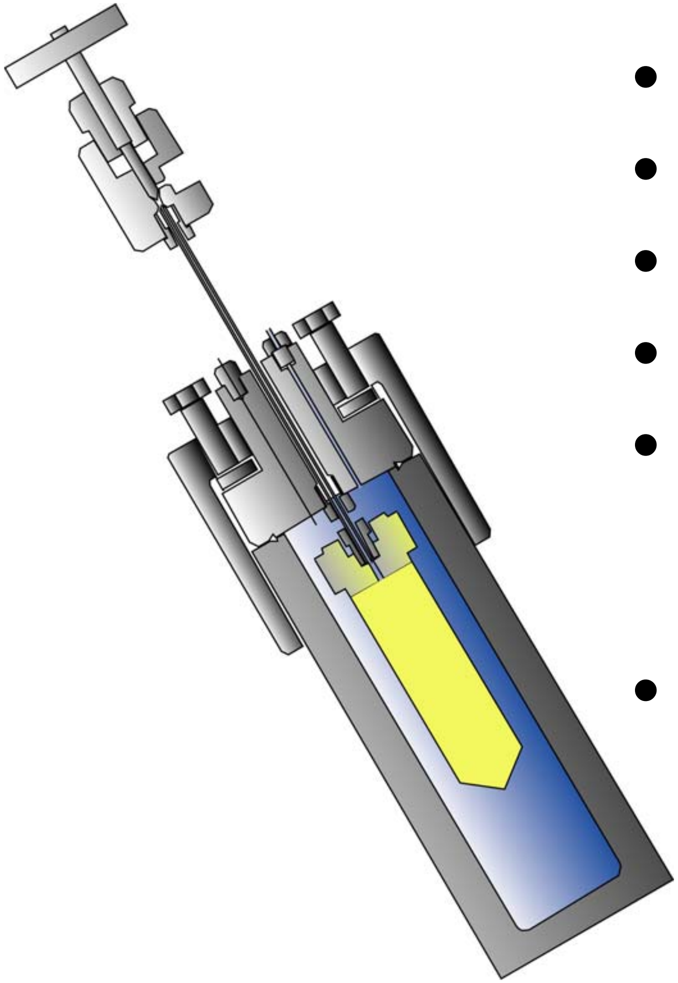
Objectives for this Study

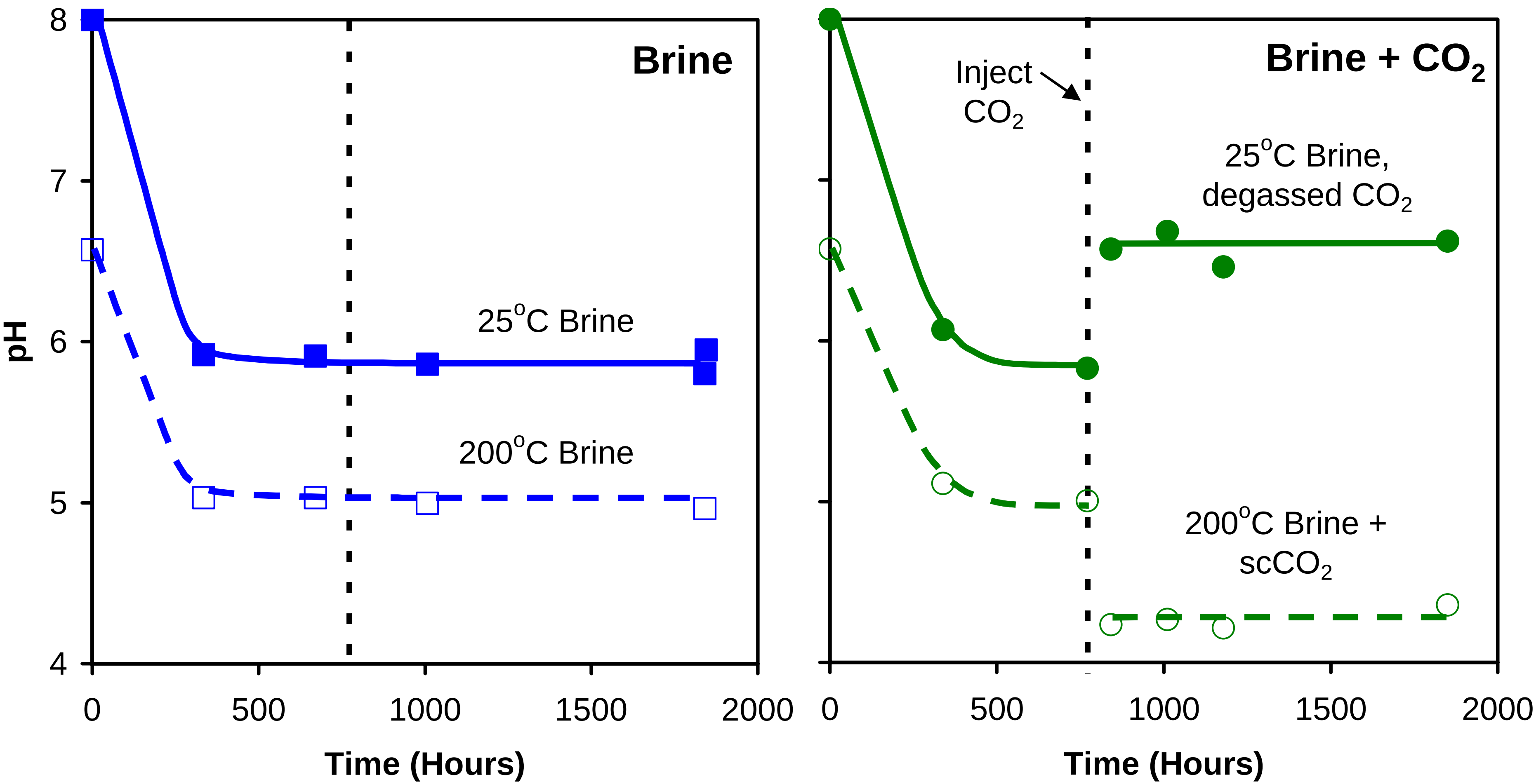
- Experimentally examine aquifer-aquitard-brine-supercritical CO_2 system that simulates geologic storage and sequestration of carbon dioxide
- Can geochemical reactions lead to failure modes in a simulated carbon repository?
- Immiscible fluids apply to a broader range of geologic processes than previously explored.
- Reactive properties of supercritical CO_2 coexisting with H_2O have implications for geochemical processes that are not understood or appreciated in the broader geologic community.

Experimental Approach

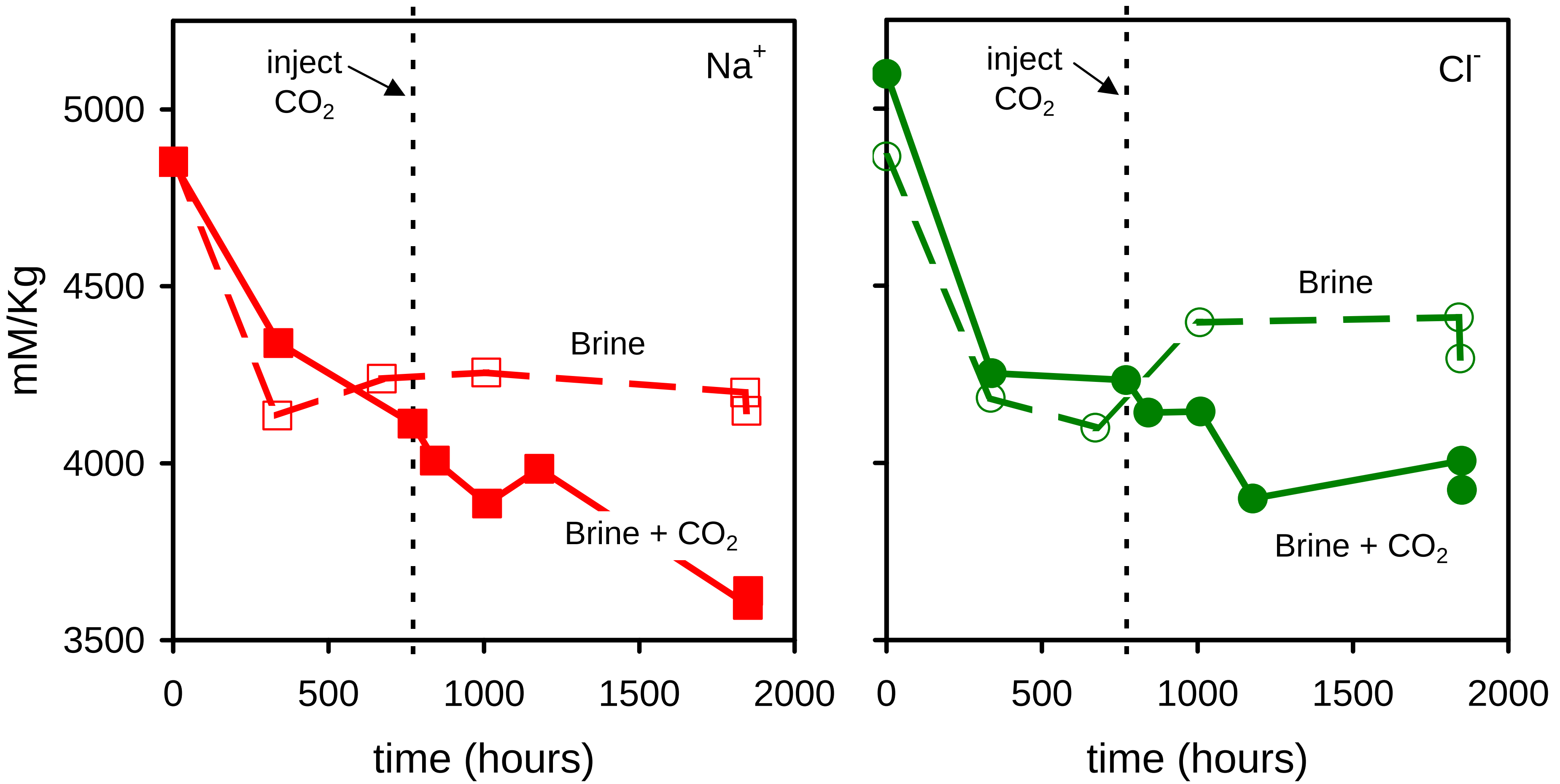


- **Flexible cell hydrothermal apparatus**
- **200°C and 200 bars**
- **5.5 molal NaCl brine**
- **Aquifer = model arkose**
- **Aquitard = argillaceous shale**
- **Experiment**
  - **Brine + rock for 32 days**
  - **Inject CO_2 into ongoing reaction, 45 days**
- **Control Experiment**
  - **Brine + rock for 77 days**

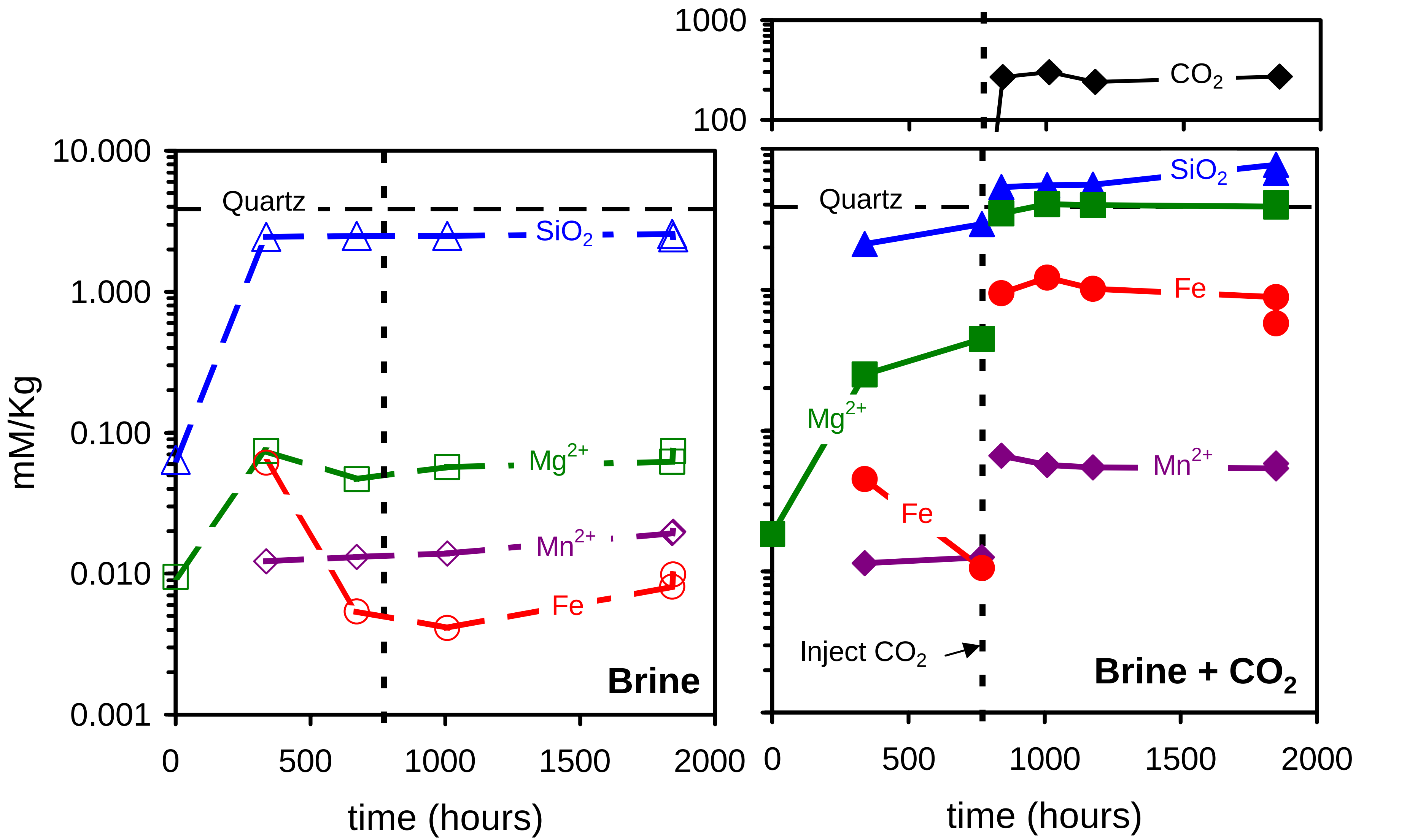
Brine Chemistry - pH



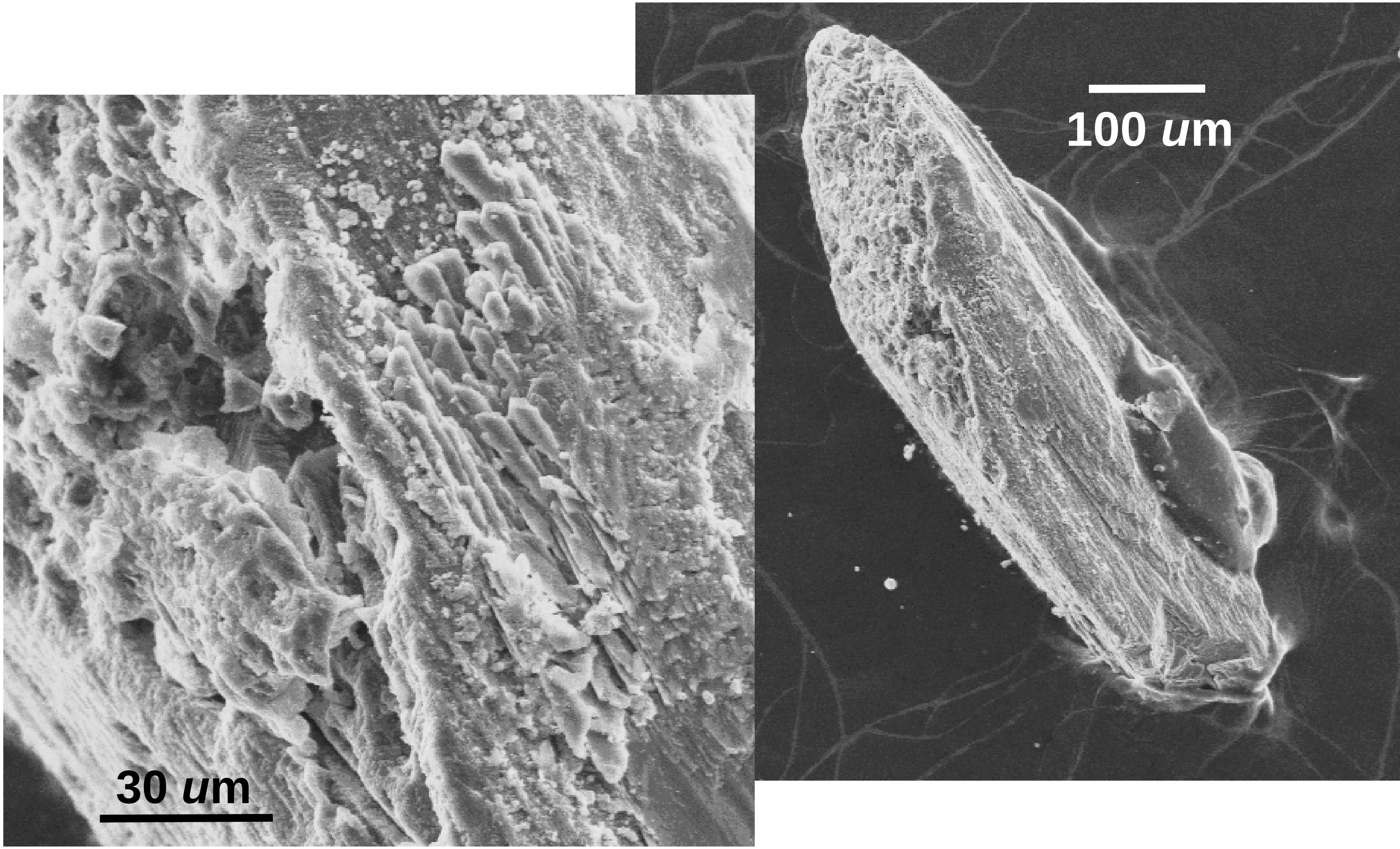
Brine Chemistry – Na and Cl



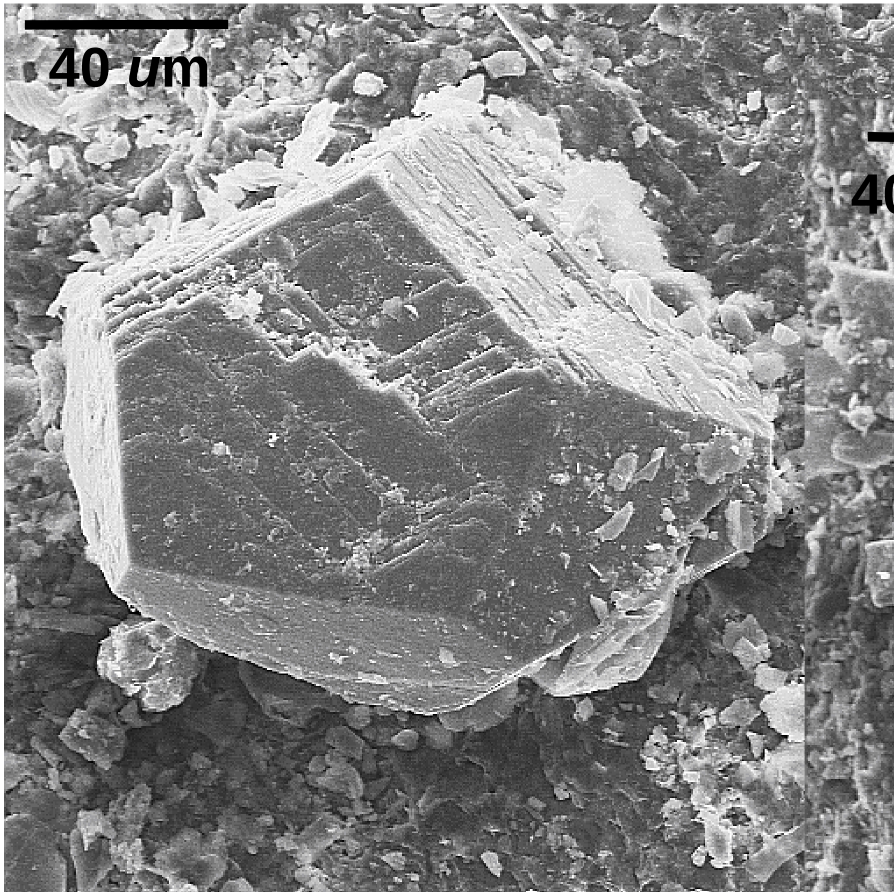
Brine Chemistry – Other Cations and CO_2

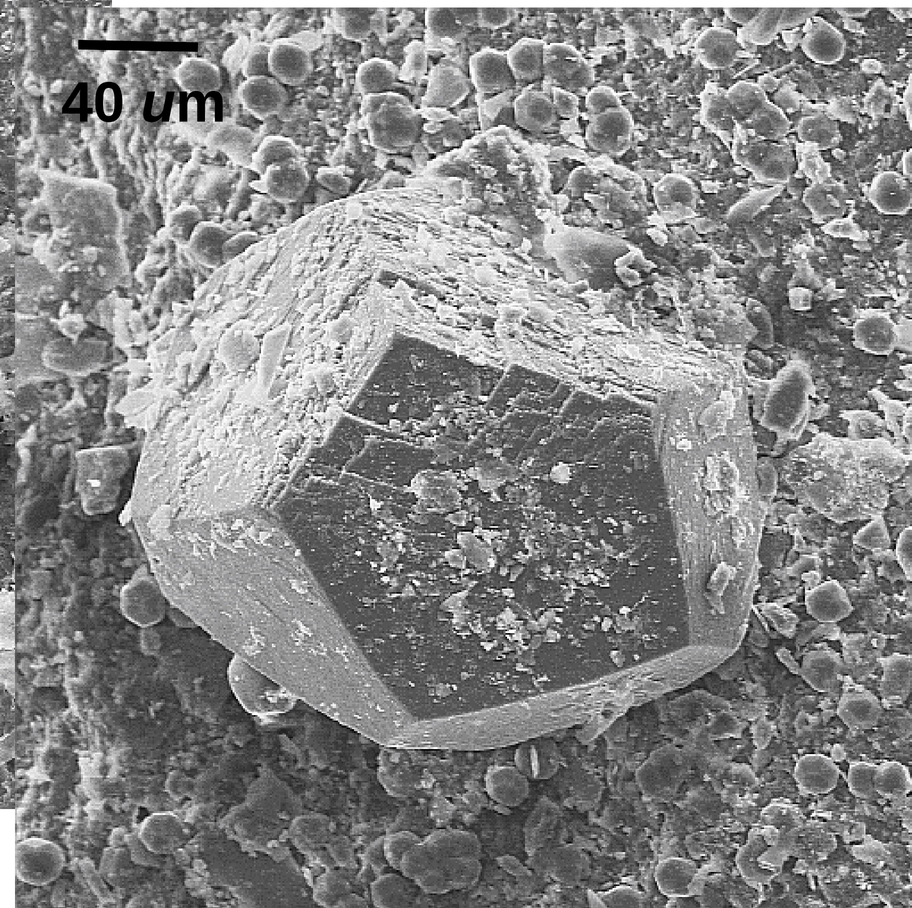


New Phase - Magnesite (CO_2-bearing Exp't)

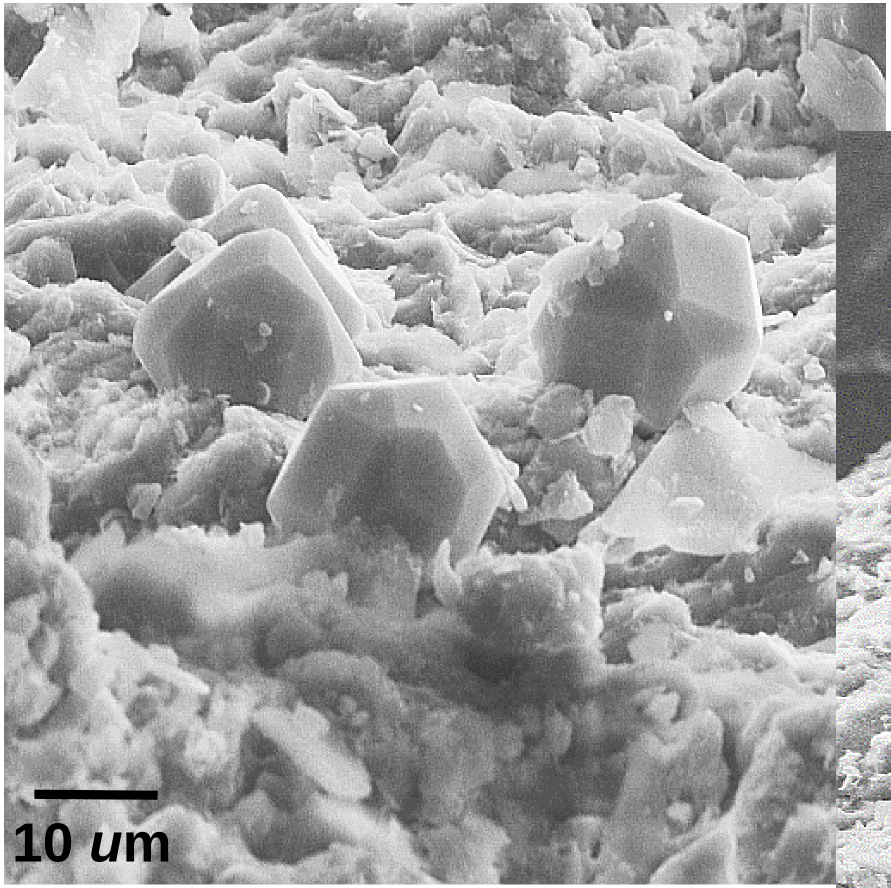


New Phase - Siderite (CO_2-bearing Experiment)



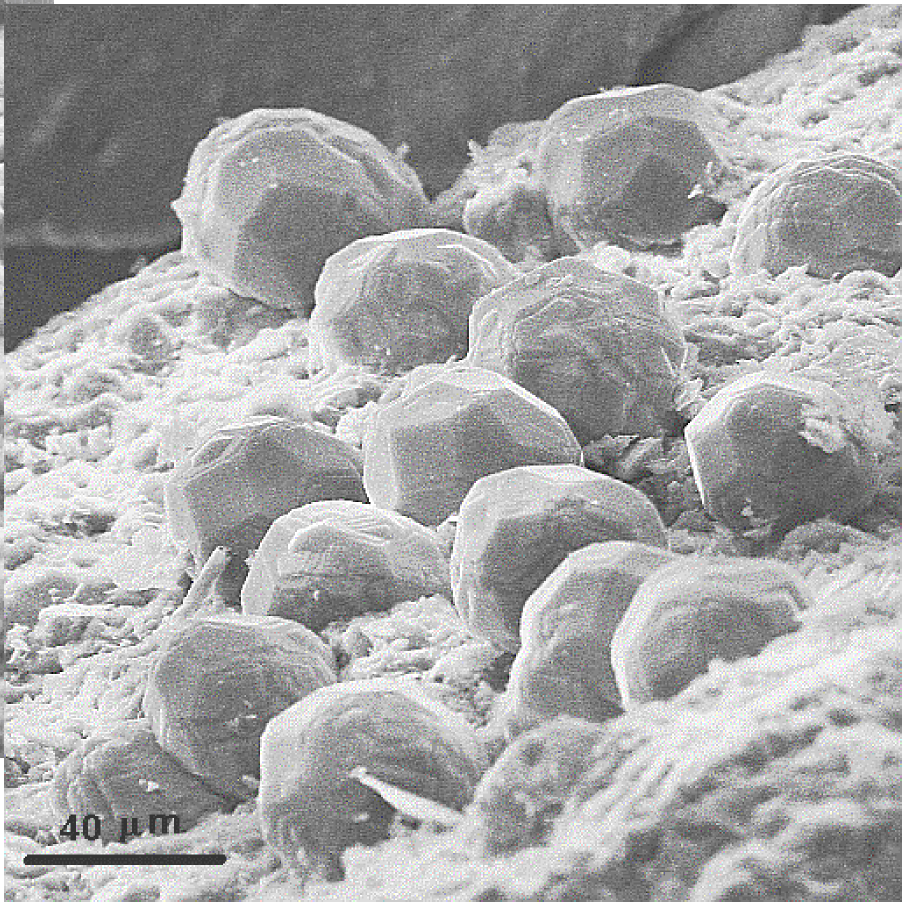


New Phase - Euhedral Analcime

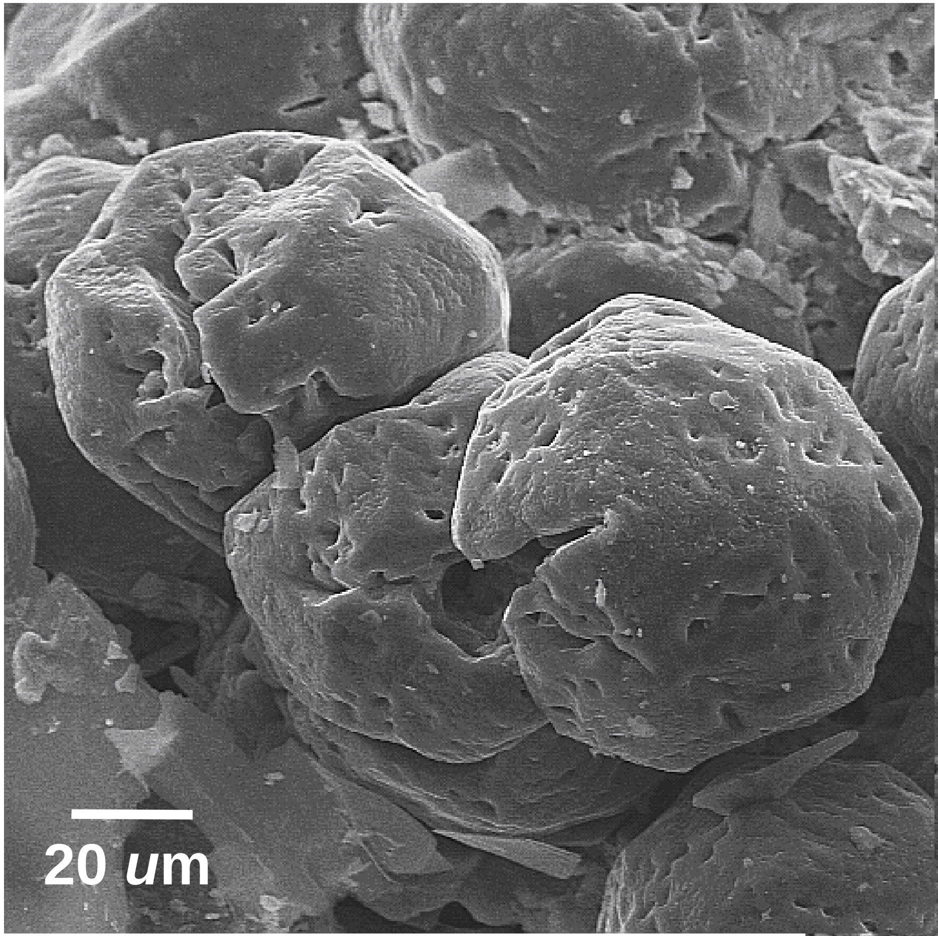


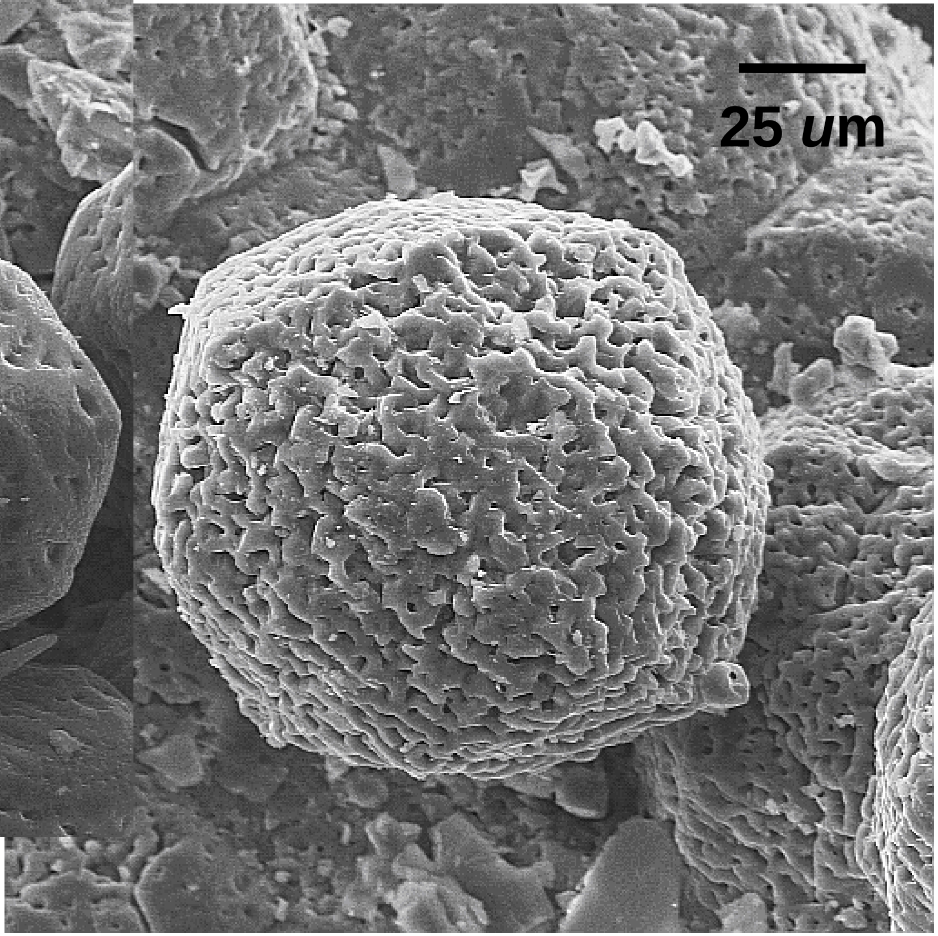
CO_2-bearing Experiment

CO_2-absent Experiment

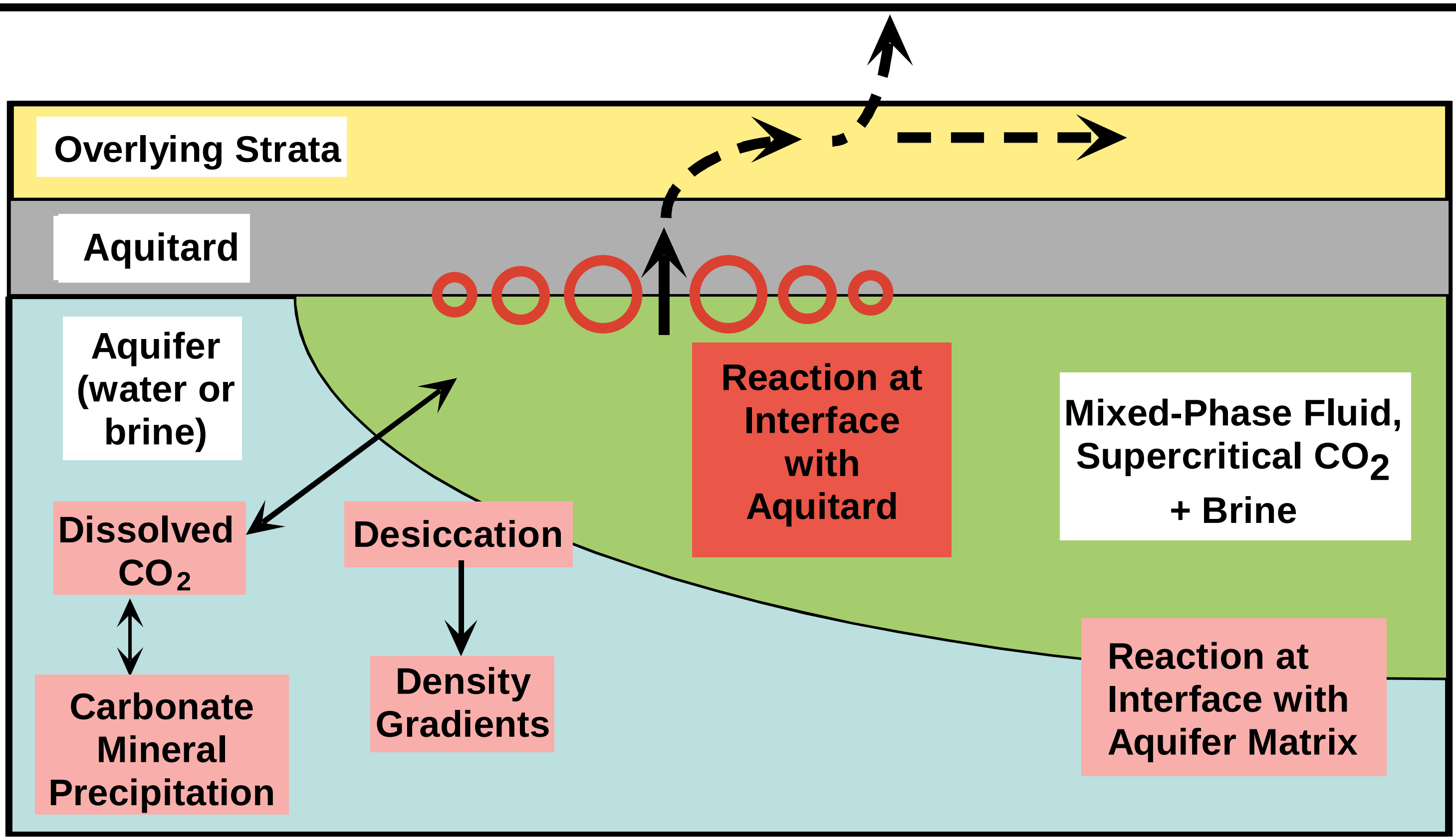


New Phase - Skeletal Analcime (CO_2-bearing)





Conclusions - Geologic Sequestration of CO_2



- ➔ Complex (non-intuitive) geochemical reaction environment. Includes immiscibility & non-carbonates. Defies simplistic assumptions & predictions
- ➔ Aquitard (shale) reacts. What coupled chemical/physical changes are important for understanding failure modes?

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Applications and Acknowledgements

- **Applications**
  - **Geologic CO_2 sequestration**
  - **Petroleum CO_2 flood recovery systems**
  - **Low-grade metamorphism**

- **Acknowledgements**
  - **US DOE BES Geoscience Program**
  - **Los Alamos National Laboratory**
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Experimental Evaluation of Mixed Fluid Reactions between Supercritical Carbon Dioxide and a NaCl Brine: Relevance to Geologic Aquifer Carbon Sequestration

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The reactive behavior of supercritical carbon dioxide under physical-chemical conditions relevant to geologic storage and sequestration of carbon is largely unknown. Experiments were conducted in a flexible cell hydrothermal apparatus to determine the extent of fluid-rock reactions, in addition to carbonate mineral precipitation, that may occur in a brine aquifer-aquitard system simulated as a saline aquifer storage scenario. The aquifer is a synthetic arkose (microcline (Or91-97)+oligoclase (An17-21)+ quartz+biotite), the aquitard is argillaceous shale, and the fluid is 5.5 molal NaCl brine. The system was held at 200 C and 200 bars for 32 days (772 hours) to approach steady state, then injected with carbon dioxide and allowed to react for an additional 45 days (1079 hours). In a separate experiment at 200 C and 200 bars, the system was allowed to react for 77 days (1845 hours) without injection of carbon dioxide.

Brine-rock reaction decreases pH from 8 to ~5.9 in both experiments. In the brine-rock-carbon dioxide experiment, injection of carbon dioxide produces a pH drop (possibly below 4, based on calculation) followed by rebound to ~6.6 as the minerals react with carbon dioxide-charged brine. Concentrations of Si and Mg in the brine increase following injection of carbon dioxide. In addition to carbonate mineral precipitation, silicate minerals (quartz, oligoclase, microcline and biotite) in the aquifer and aquitard display textures (etch pits, mineralization) indicating significant reactions. A pressure decrease of 23 bars occurred in the experimental cell over a 3-day period following carbon dioxide injection. Pressure was stable afterwards. The pressure decrease is interpreted as consumption of supercritical carbon dioxide by dissolution in brine and subsequent precipitation as carbonate mineral. The experimental reactions provide initial constraints on reactions and reaction rates that can impact the containment interface in moderate temperature brine aquifer systems with potential for carbon sequestration.

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